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First real-time measurements of N_2O isotopic signatures above intensively managed grassland: analytical performance, validation and illustrative examples

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Measurement of the four main N_2O isotopic species ($^{14}N^{15}N^{16}O$ / $^{15}N^{14}N^{16}O$ / $^{14}N^{14}N^{18}O$ / $^{14}N^{14}N^{16}O$) has been suggested as a powerful tool to trace the biogeochemical cycle of N_2O and to allocate its emission sources. Studies carried out with microbial pure cultures and mixed population systems (Wunderlin et al. 2012) allowed the determination of characteristic isotopic signatures for the most important production processes. These characteristic signatures have been applied to identify relevant sources at different scales (Park et al. 2012). However, current studies suffer from limited spatial and temporal resolution due to the combination of discrete flask sampling in conjunction with laboratory-based mass spectrometric analysis. We recently demonstrated that a quantum cascade laser (QCL) based absorption spectrometer is capable of simultaneously measuring the three main N_2O isotopomers at trace levels (Waechter et al. 2008). Furthermore, its potential for in-situ measurements in conjunction with a liquid nitrogen-free preconcentration unit has been proven (Mohn et al. 2012).

Here we present results from the first long-term field measurement campaign conducted on intensively managed grassland in central Switzerland during three months. A modified state-of-the-art laser spectrometer (Aerodyne Research, Inc.) employing a mid-infrared cw-QCL (4.54 μm) and a novel astigmatic multipass cell with 204 m optical path-length was connected to a N_2O preconcentration unit. High analytical performance in combination with the applied calibration strategy resulted in excellent long-term precision of 0.20, 0.12 and 0.11‰ for $\delta^{15}N^{\alpha}$, $\delta^{15}N^{\beta}$ and $\delta^{18}O$ which was determined from repeated preconcentration and measurement of target gas from a compressed air tank. This instrumental setup allowed investigating responses of isotopic composition in soil-emitted N_2O to management events and weather influences. The accompanying measurements of soil temperature, soil water content, ammonia, and nitrate concentrations made the identification of controls on N_2O isotopic composition possible. Furthermore, simultaneous eddy-covariance N_2O flux measurements (Merbold et al. 2014) were used to derive a flux-averaged isotopic signature of soil-emitted N_2O of intensively managed grassland. In this context, the potential of the derived N_2O isotopic signatures for partitioning of microbial source processes will be discussed in relation to available literature data.

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